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INFORMATION DESIRED BY

I. URANIUM LOSSES FROM THE Y-12 PLANT TO THE ENVIRONMENT

An analysis has been made of the losses of uranium from the Y-12 Plant during the period August, 1943 to May, 1956. It was necessary to estimate most of the losses from operating experience and the few exhaust-gas analysis data which were available. The best known losses were the discharges to the sewer which flows into Poplar Creek; however, these should only be regarded as an estimate with good reliability. For the estimation of these losses, uranium accountability and operating records have been utilized as a basis. The losses are tabulated in Table I. A description of the effluents is outlined as follows:

The Alpha Process, 1943 to 1945

The separation of the isotopes of uranium by the electromagnetic process involved several chemical operations in addition to the physical process. The separation itself was only about 7-8% efficient. This meant that most of the uranium had to be recovered and made into new charge material.

The Calutron units were rather complicated structures of metal, largely copper, carbon and ceramics. After use, the various parts of the machine were washed in dilute nitric acid solution. Since the charge material to the unit was uranium tetrachloride, the washing generated a rather corrosive solution from which the uranium had to be recovered and remade into charge material. Various processes were used at different times, but most of the material was recovered by one in which the iron was removed from the solutions by pH adjustment and filtration. The uranium was precipitated as the peroxide and the solution was filtered. The filtrates from the peroxide step contained a fair amount of uranium which passed to the

which there were fairly large dust losses. The oxide was converted to the chloride by heating with liquid carbon tetrachloride under pressure or by a vapor phase process. The chloride was further purified by sublimation when required. From most of the operations the losses were in the form of solutions. They were drained by the sewer into Poplar Creek. It is estimated that the air-borne losses could not have been more than a few percent of the total losses from the process. The uranium from carbon parts was recovered by burning the carbon. There was also some dust loss from this process.

The Beta Process, 1944 to 1947

The purpose of the Beta cycle was to concentrate the product of the Alpha cycle. Because of the enhanced value of the material, all of the operations were much more carefully done. The Beta Calutron units were smaller and more efficient. The chemical recycle, while it had the same objective of reconversion of the unused material to new uranium tetrachloride for

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reuse, was done in a more quantitative manner. The effluents from the various precipitation steps were recovered by solvent extraction and more careful attention was paid to the prevention of dust losses. Even though the total losses were much lower, it is possible that the dust losses were a somewhat higher percentage of the total losses than was the case in the Alpha process.

### Product Processing

#### A. Building 9206, Beta Product Processing, 1944 to 1947

The Beta product was collected on carbon parts. These were cleaned of the bulk of the material, which was in the form of a loose powder, and the remaining carbon was burned to insure complete recovery. Because the input to the cycle had to be measured after several processing steps had taken place, the actual losses in this cycle were in some doubt; however, all of the operations were done with great care and the losses are estimated to be low. The collected material was put into nitric acid solution, purified by ether extraction, precipitated as the peroxide, converted to the oxides and made to uranium tetrafluoride with hydrogen and hydrogen fluoride. It is estimated that there were dust losses in these dry operations and they may have been a sizeable percentage of the total losses from the system. Undoubtedly some of the dust losses were never detected in the product cycle and, as a consequence, would have appeared as losses in the Beta separation process.

#### B. Building 9212

##### 1. Process Vents from Uranium Hexafluoride Reduction, D-Wing, 1954 to Date

The continuous process for reducing uranium hexafluoride to uranium tetrafluoride with hydrogen and fluorine had a small amount of unreacted uranium hexafluoride or uranium tetrafluoride dust remaining in the vent gases after the process gas passed through mechanical filters and chemical traps. The process gas was discharged continually to the atmosphere. Any unreacted uranium hexafluoride would have been immediately hydrolyzed to uranyl fluoride smoke upon contact with moist air.

##### 2. Process Vents from Uranium Tetrafluoride Preparation by Batch Hydrofluorination, 1945 to Date

The production of uranium tetrafluoride by this process involved precipitating water solutions of uranium salts with peroxide and reducing the cake with hydrogen batchwise to the oxide. The oxide was fluorinated batchwise with gaseous hydrogen fluoride at elevated temperatures. Dusting was inhibited by passing the vent gases through a porous carbon filter and a neutralizing pot before venting to an exhaust air stack. The effluents from this process were probably as fumes or dust from the reduction and fluorination steps.

3. Discards from Chemical Processing to Storm Sewer, 1953 to Date

These discards consisted primarily of dilute uranium solutions which were uneconomical to salvage and were usually basic in nature. They also contained appreciable amounts of nitrates and fluorides. This sewer emptied into Poplar Creek.

4. Airborne Ventilation Losses from Product Processing, 1945 to Date

These airborne losses were discharged principally as uranium oxide, uranyl fluoride and uranyl nitrate fumes, or uranium tetrafluoride dust. Very little information can be furnished about the relative amounts of each which were discharged. They left the building diluted with large volumes of exhaust air.

5. Losses Outside the Plant Area on Shoes and Clothing, 1945 to Date

The losses were carried from the plant in small amounts by personnel who occasionally needed to be in the work areas but were not exposed sufficiently to contamination to warrant protective clothing. The uranium consisted primarily of oxide and nitrate.

Intermediate Assay Processing

A. Process Vents from Uranium Tetrafluoride Preparation, 1955 to Date

The losses occurred in a manner similar to that of the 9212 process vent losses described in paragraph B-1 above, but the material was of lower assay.

B. Airborne Ventilation Losses, 1955 to Date

These losses occurred in a manner similar to that of the 9212 ventilation losses described in paragraph B-4 above.

C. Airborne Salvage Area Losses, 1946 to Date

These losses occurred primarily as dust or fumes in the form of uranyl nitrate, uranium tetrafluoride, urano-uranyl oxide, uranium trioxide and uranyl fluoride diluted with large volumes of exhaust air.

D. Discards from Chemical Processing to Storm Sewer, 1946 to Date

The discards consisted of solutions from which the dissolved uranium could not be recovered economically. These solutions contained uranium in many forms; for example, uranyl nitrate, ammonium di-uranate and uranium oxides.

E. Accidental Uranium Hexafluoride Release to the Atmosphere

On May 11, 1956, the hydrogen line to the uranium hexafluoride tower broke. This caused incomplete conversion of the uranium hexafluoride to uranium tetrafluoride. The chemical traps which were provided to recover the uranium hexafluoride in such an emergency were filled to capacity before the situation was corrected, and uranium hexafluoride was allowed to escape to the atmosphere. The escaping uranium was visible as uranyl fluoride smoke as it was emitted from the vent stack.

F. Losses Outside the Plant Area on Shoes and Clothing, 1946 to Date

These losses occurred in the same manner as those described in the preceding section, paragraph B-5, but were smaller because the amount of traffic and the throughput were less.

Normal and Depleted Assay Processing

A. Building 9212

1. Airborne Ventilation and Hood Discharge, 1948 to 1954

These losses were primarily in the form of uranium oxide and uranium metal dust from the Sunflower foundry and machining operations. No filters were provided prior to September, 1953. The losses for the period were estimated from unaccounted-for material figures.

2. Airborne Ventilation and Hood Discharge, 1954 to Date

The dust was in the same form as that from 1948 to 1954, but the losses were larger. The production level increased and the filters which were installed in 1953 required excessive maintenance. In the Spring of 1955 extensive repair of the filters was made and essentially uninterrupted operation has been maintained since July 1, 1955. From January, 1954 to July, 1955, there were probably large losses since little dust was collected compared with the amount now being recovered. The exact amount can only be estimated from present recovery figures, making adjustments for the level of production. The losses during FY-1956 have been estimated from an analysis of stack gas composition to be about 0.1 kg. per day. A continuous monitor is now being installed on the exhaust stack.

B. Building 9206

1. Combustible Salvage Burning, 1948 to 1955

These losses were primarily airborne uranium oxide resulting from burning combustible salvage which, prior to December, 1955, consisted of a mixture of uranium metal and oxide from the 9212 casting operation. This operation has not been conducted in the Y-12 Plant since 1955.

2. Discard of Salvage to Sewer, 1946 to Date

This salvage was similar in nature to that described in the preceding section, paragraph D.

3. Chip Crushing Flush to Sewer, 1955 to Date

The chips from the machining operation were flushed with water while being crushed prior to briquetting. Most of this loss, in which the uranium was in the form of the oxide, has occurred in the last six months.

C. Building 9211

1. Airborne Salvage Burning and Kiln Losses, 1953 to Date

Combustible organic-uranium salvage was burned in a Herreshoff furnace prior to leaching. The exhaust gases from the furnace contained uranium trioxide and urano-uranic oxide dust. The ash was leached and precipitated as ammonium diuranate. This was calcined to the oxide in an indirectly-fired rotary kiln. Considerable dust losses occurred from this operation. In December, 1955, a scrubber was put into operation, reducing the losses from both operations to about 0.3 kg./day when the salvage burning was being carried out.

2. Airborne Ventilation Losses, 1953 to Date

These airborne losses were primarily uranyl nitrate, uranium oxides, and ammonium diuranate, derived from the processing of old salvage which had been in storage.

3. Discards to Sewer, 1953 to Date

These discards were similar to those described in the preceding section, paragraph D.

Discards to Disposal Pits and Burial Grounds, 1947 to Date

The estimate of the amount of uranium discarded to the burial grounds and disposal pits was based on uranium accountability and operating records. Most of the material was discarded in recent years. The uranium was in numerous chemical forms, among these being uranium oxides, uranium nitrate, ammonium diuranate and uranium peroxide solutions.

TABLE I

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## URANIUM LOSSES FROM THE Y-12 PLANT TO THE ENVIRONMENT

Source	Period	Uranium, kg.	Basis for Estimate	Isotopic Composition, Percent		
				U-234	U-235	U-236
Alpha Process	1943 to 1945	40,000	Estimated from known losses and unaccounted-for material	0.005	0.65	0
Beta Process	1944 to 1947	400	Estimated from known losses and unaccounted-for material	0.7	23.5	0
Product Processing						
A. <u>Building 9206, Beta Product</u>	1944 to 1947	0.4	Estimated from unaccounted-for material	1	80	0
B. <u>Building 9212</u>						
1. Process Vents from UF <sub>6</sub> Reduction, D-Wing	1954 to Date	0.3	Estimated from a study of the composition of vent gases, and operating experience.	1	93	0.33
2. Process Vents from UF <sub>6</sub> Preparation by Batch Process	1945 to 1952 1953 to Date	0.5 1.5	Estimated from a study of the composition of vent gases.	1 1	93 93	0. to 0.1 0.2 to 1.0
3. Discards from Chemical Processing to Sewer	1953 to Date	5.2	Accountability and operating records used as basis	1	93	0.4
4. Airborne Ventilation Losses from Product Processing	1945 to 1952 1953 to Date	1.7 1.7	Estimated from an analysis of air sample data.	1 1	93 93	0 to 0.1 0.2 to 0.5
5. Losses Outside Plant Area on Shoes and Clothing	1945 to Date	1	Estimated as order of magnitude	1	93	0 to 1.0
Intermediate Assay Processing, Building 9206						
A. Process Vents from UF <sub>6</sub> Preparation	1955 to Date	0.1	Estimated from a study of the composition of vent gases, and operating experience.	0.2	37.5	0.31
B. Airborne Ventilation Losses	1955 to Date	0.5	Estimated from operating experience in similar operations	0.2	37.5	0.31
C. Airborne Salvage Area Losses	1946 to 1952 1953 to Date	0.8 5.2	Estimated	0 to 0.1 0.2	2 25	0 to 0.25 0.2
D. Discards from Chemical Processing to Storm Sewer	1946 to 1952 1953 to Date	2.1 13.2	Accountability records used as basis	0 to 0.1 0.2	2 25	0 to 0.25 0.2
E. Accidental UF <sub>6</sub> Release to the Atmosphere	5/11/56	2	Measured by difference after clean-up	0.2	37.5	0.36
F. Losses Outside Plant Area on Shoes and Clothing	1946 to Date	0.5	Estimated as an order-of magnitude	0.4	20	0 to 0.3
Normal and Depleted Assay Processing						
A. <u>Building 9212</u>						
1. Airborne Ventilation and Hood Discharge	1948 to 1954	1,500	Estimated from unaccounted-for losses	0.002 to 0.005	0.4 to 0.7	0 to 0.01
2. Airborne Ventilation and Hood Discharge	1954 to Date	5,000 to 10,000	Estimated from present recovery figures. (Loss since 7/1/55 is approximately 0.1 kg./day)	0.002	0.2 to 0.4	0.01
B. <u>Building 9206</u>						
1. Combustible Salvage Burning	1948 to 1955	3,200	Estimated from operating experience	0.002 to 0.005	0.15 to 0.7	0 to 0.01
2. Discards from Salvage to Sewer	1946 to Date	1,435	Accountability records used as basis.	0.005	0.6	0 to 0.01
3. Chip Crushing Flush to Sewer	1955 to Date	60	Estimated from operating experience	0.002	0.2 to 0.4	0.01
C. <u>Building 9211</u>						
1. Airborne Salvage Burning and Kiln Losses	1953 to Date	120	Estimated to Dec. 1955; calculated from effluent analysis Dec. 1955 to Date.	0.002 to 0.015	0.2 to 0.8	0.01
2. Airborne Ventilation Losses	1953 to Date	10	Estimated from operating experience.	0.002 to 0.015	0.2 to 0.8	0.01
3. Discards to Sewer	1953 to Date	900	Accountability records used as basis	0.002 to 0.015	0.6 Avg.	0.01
Discards to Disposal Pits and Burial Grounds	1943 to Date	9,000	Accountability and operating records used as basis.	0.002 to 1.0	0.1 to 93 Avg. 0.4	0 to 0.40

## II. ISOTOPIC COMPOSITION OF ORGDP PRODUCT AND TAILS

Isotopic assays for Oak Ridge Gaseous Diffusion Plant product from September, 1945 through May, 1956 are presented in table II. The uranium-235 percentages are based on analyses of cascade semi-monthly inventory samples, the uranium-234 and uranium-236 on an average of results from samples taken each eight-hour shift.

Uranium-235 assays for tails materials from June, 1946 through May, 1956 are given in table III. The values prior to 1953 are based on analyses of cascade semi-monthly inventory samples, those from 1953 to the present on an average of results from samples taken each eight-hour shift. The available uranium-234 and uranium-236 assays for tails materials are shown in table IV. Each value in this table represents a single sample.



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TABLE II

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## ISOTOPIC COMPOSITION OF ORGDP PRODUCT, WEIGHT PERCENT

	JAN.	FEB.	MAR.	APR.	MAY	JUNE	JULY	AUG.	SEPT.	OCT.	NOV.	DEC.
<u>1945</u>												
U-235	-	-	-	-	-	-	-	-	46.30	54.80	59.10	58.20
<u>1946</u>												
U-235	56.01	50.18	50.00	48.50	50.00	60.08	50.08	60.07	50.03	60.04	60.05	93.47
<u>1947</u>												
U-234	1.56*	1.45*	1.30*	1.21*	1.13*	1.08*	1.07*	1.09*	1.10*	1.10*	1.17	1.17
U-235	93.42	93.52	93.53	93.56	93.41	93.36	93.35	-	94.54	94.54	93.09	93.55
U-238	5.02	5.03	5.17	5.23	5.46	5.56	5.58	-	4.36	4.36	5.74	5.28
<u>1948</u>												
U-234	1.16	1.14	1.13	1.13	1.16	1.16	1.21	1.18	1.15	1.13	1.11	1.12
U-235	93.19	93.23	93.12	93.24	93.20	93.26	93.15	93.08	93.32	93.21	93.18	93.15
U-238	5.65	5.63	5.75	5.63	5.64	5.58	5.64	5.74	5.53	5.66	5.71	5.73
<u>1949</u>												
U-234	1.13	1.12	1.13	1.12	1.11	1.08	1.07	1.07	1.10	1.10	1.09	1.09
U-235	93.13	93.20	93.22	93.20	93.31	93.25	93.18	93.27	93.20	93.25	93.21	93.20
U-238	5.74	5.68	5.65	5.68	5.58	5.67	5.75	5.66	5.70	5.65	5.70	5.71
<u>1950</u>												
U-234	1.09	1.08	1.07	1.06	1.08	1.06	1.21	1.35	1.24	1.16	1.13	1.12
U-235	93.24	93.40	93.32	93.22	93.20	93.26	93.08	92.85	92.70	93.19	93.15	93.26
U-238	5.67	5.52	5.61	5.72	5.72	5.68	5.71	5.80	6.06	5.65	5.72	5.62
<u>1951</u>												
U-234	1.15	1.19	1.24	1.22	1.17	1.11	1.07	1.06	1.08	1.11	1.16	1.17
U-235	93.15	93.17	93.26	93.11	93.12	93.16	93.14	93.18	93.20	93.17	93.15	93.12
U-238	5.70	5.64	5.50	5.67	5.71	5.73	5.79	5.76	5.72	5.72	5.69	5.71
<u>1952</u>												
U-234	1.17	1.14	1.10	1.07	1.05	1.02	1.00	0.97	0.97	0.97	1.00	1.00
U-235	93.20	93.06	93.04	93.12	93.25	93.16	93.09	93.14	93.08	93.15	93.10	93.26
U-238	5.63	5.80	5.86	5.81	5.70	5.82	5.91	5.89	5.95	5.88	5.90	5.74
<u>1953</u>												
U-234	1.03	1.07	1.10	1.14	1.19	1.21	1.21	1.20	1.18	1.16	1.13	1.10
U-235	93.21	93.19	93.17	93.30	93.20	93.29	93.30	93.35	93.30	93.34	93.31	93.16
U-236	-	-	-	-	-	0.13	0.18	0.19	0.22	0.23	0.25	0.26
U-238	5.76	5.74	5.73	5.56	5.61	5.37	5.31	5.26	5.30	5.27	5.31	5.48
<u>1954</u>												
U-234	1.10	1.10	1.08	1.05	1.04	1.03	1.03	1.02	1.02	1.02	1.00	0.96
U-235	93.20	93.28	93.21	93.22	93.20	93.18	93.16	93.18	93.18	93.14	93.16	93.42
U-236	0.27	0.28	0.27	0.28	0.27	0.29	0.30	0.31	0.31	0.32	0.32	0.32
U-238	5.43	5.34	5.44	5.45	5.49	5.50	5.51	5.40	5.49	5.52	5.52	5.30
<u>1955</u>												
U-234	0.95	0.99	1.01	0.97	0.97	1.04	1.08	1.07	1.07	1.07	1.05	1.02
U-235	93.45	93.75	93.76	93.42	93.27	93.58	93.46	93.42	93.21	93.19	93.20	93.17
U-236	0.34	0.33	0.32	0.32	0.32	0.30	0.28	0.28	0.29	0.31	0.31	0.32
U-238	5.26	4.93	4.91	5.29	5.44	5.03	5.18	5.23	5.43	5.43	5.44	5.49
<u>1956</u>												
U-234	0.98	0.91	0.82	0.81	0.80							
U-235	93.23	93.27	93.27	93.30	93.33							
U-236	0.31	0.32	0.35	0.40	0.42							
U-238	5.48	5.50	5.56	5.49	5.45							

\* By alpha counting.

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TABLE III

WEIGHT PERCENT URANIUM-235 IN ORGDP TAILS

	1946	1947	1948	1949	1950	1951	1952	1953	1954	1955	1956
January	-	0.456	0.510	0.522	0.482	0.494	0.440	0.552	0.949	0.414	0.279
February	-	0.499	0.512	0.509	0.494	0.479	0.445	0.582	0.962	0.412	0.247
March	-	0.484	0.519	0.502	0.479	0.498	0.428	0.654	0.932	0.386	0.228
April	-	0.487	0.508	0.502	0.493	0.479	0.444	0.638	0.885	0.419	0.252
May	-	0.478	0.512	0.505	0.486	0.471	0.438	0.664	0.783	0.400	0.299
June	0.494	0.512	0.509	0.502	0.487	0.462	0.431	0.700	0.760	0.355	
July	0.526	0.521	0.508	0.511	0.485	0.490	0.418	0.727	0.635	0.435	
August	0.489	0.520	0.506	0.502	0.547	0.476	0.428	0.773	0.555	0.350	
September	0.503	0.531	0.507	0.514	0.555	0.440	0.415	0.863	0.559	0.451	
October	0.491	0.510	0.514	0.498	0.565	0.450	0.395	0.879	0.514	0.396	
November	0.478	0.506	0.490	0.482	0.533	0.446	0.470	0.948	0.479	0.350	
December	0.454	0.520	0.505	0.514	0.500	0.440	0.475	0.956	0.406	0.310	

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TABLE IV  
CONCENTRATION OF MINOR ISOTOPES IN ORGDP TAILS

Date	Composition, Weight Percent		
	U-234		U-236
	Alpha Counting	Spectrometer	
<u>1947</u>			
January	0.0034		
March	0.0026		
April	0.0026		
May	0.0028		
June	0.0027		
July	0.0030		
August	0.0030		
September	0.0031		
November	0.0030		
December	0.0029		
<u>1948</u>			
January	0.0028		
February	0.0028		
<u>1953</u>			
April	0.0044		
<u>1954</u>			
March	0.0058		
<u>1955</u>			
May		0.0025	0.0075
<u>1956</u>			
April		0.0025	0.0070